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A VACUUM-SPRAY TECHNIQUE FOR THE ATOMIZATION OF LIQUID METALS

by

K.G. DAVIS*, C.F. DIXON† AND J. G. MAGNY**

ABSTRACT

A vacuum-spray atomization device for the production of metal powder with low gas content has been built and tested. Molten metal was passed through a nozzle into a vacuum chamber, with argon gas introduced into the nozzle stream. Expansion of the gas on entering the vacuum chamber provided the energy for atomization. Wood's metal was successfully sprayed in a low-temperature model. A similar unit, designed for use with higher-melting-point metals, was then made. Cast iron was chosen as a prototype material for spraying. Much coarser powder was obtained. Based on a series of tests with various nozzle designs, it was concluded that the process would only work for steels or superalloys if a nozzle-heater were introduced and the metal reservoir were pressurized.

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INTRODUCTION

In accord with the CANMET objective "to develop and promote the technological base for improving the quality of materials for the continued expansion of Canadian industry", work has been done at PMRL on a procedure for the vacuum-atomization of liquid metals. If successful, such a procedure could be utilized by the Canadian powder metals industry for the production of alloy powders. Canada is already one of the world's leading producers of iron powder.

In conventional atomization of liquid metals, a stream of liquid emerging from a nozzle is struck by jets of water or gas. The metal is both atomized and quenched, and falls as a powder to the base of the atomization chamber. The use of water jets is unsuited to applications where oxidation of the powder is undesirable, and even inert gas jets can cause some oxidation unless gas of very high purity is used. For certain superalloys, oxidation of the powder-particle surfaces must be avoided for optimum properties after sintering, and some manufacturers do not allow air to contact the powder at any stage prior to compaction.

Wentzell(1) has described a process in which a soluble gas is introduced under pressure into a reservoir of molten alloy, the alloy then being fed through a nozzle into an evacuated chamber. Evolution of the dissolved gas from the molten metal stream as it enters the vacuum chamber is sufficiently energetic to cause atomization of the metal. N_2, H_2, and CO, have all been used successfully. The technique gives a powder with low gas content and a clean surface, and uses only moderate amounts of gas. The process is, however, inherently complex. The alloy must be kept molten under pressure while the gas is absorbed into the melt, which necessitates a pressure-tight furnace design. Further, H_2 is the preferred gas for absorption, and the use of hydrogen under pressure at high temperatures and its subsequent pumping makes the conscientious

adherence to safety procedures mandatory. Finally, there is no gas or water jet to quench the molten metal droplets, which must freeze while travelling in a vacuum. Consequently, large vacuum chambers, typically 15 m in height, have to be used.

Olsson and Turkdogan(2) studied the effect of the introduction of argon gas bubbles into the nozzle stream when vacuum-degassing steel. They showed that the bubbles led to "a radial spread of the stream, and spray formation". Degassing efficiency was improved. In the current work, this concept has been explored as a method for producing powder. It depends for success on the energy of the expanding gas bubbles as they enter the vacuum being sufficient to atomize the liquid metal. The projected method will be far simpler than that of Wentzell. A standard melting method is used, followed by pouring into a preheated tundish set on a vacuum chamber, the nozzle at the tundish base being modified to allow the introduction of gas. The gas will provide some cooling, which will allow a smaller vacuum chamber to be used than that for the Wentzell process.

The principle aim of the project is to develop a relatively simple vacuum-spray method to produce alloy powders with good surface quality and low gas content.

PLEXIGLASS/WOOD'S METAL MODEL SYSTEM

Apparatus

To test the feasibility of the technique and to examine nozzle design, a system with a plexiglass tundish and nozzle was built, see Fig. 1. Wood's metal (50% Bi, 27% Pb, 13% Sn, 10% Cd), with a melting point around 70°C, was chosen as the alloy to be atomized. Two nozzle designs were initially tested, shown in Fig. 2. Design (b), where the gas was introduced around the periphery of the nozzle exit, never gave powder, and will not be discussed further. The runs were all done, therefore, on variations of design (a).

Nozzle diameter was always 4.8 mm. The nozzles with 16 gas inlet holes had two sets of 8 holes at different levels in the nozzle. In those with 8 holes or less, the holes were all on the same level. In the nozzles with two gas inlets, the holes were diametrically opposite one another. It was found early in these runs that some improvement in the spray geometry was obtained if the nozzle was extended, which was done by attaching to the nozzle base a piece of copper tubing with inner diameter equal to that of the nozzle. The following procedure was followed. The nozzle entrance was closed off with a stopper rod and the spray chamber was pumped down to less than 1 mm Hg. Argon gas was then introduced into the nozzle manifold and the flow adjusted to give the desired manifold pressure. This pressure will hereafter be referred to as the 'initial' manifold pressure, to differentiate from the manifold pressure during the spray. Pumping capacity of the vacuum system was sufficiently large that the gas flow caused no measurable loss of vacuum in the spray chamber. When the required gas flow had been established, molten Wood's metal at around 100°C was poured into the container above the nozzle, and the stopper rod removed. Spray time was noted with a stop watch. The manifold pressure rose above atmospheric during spraying. It was found that initial manifold pressures greater than about 700 mm Hg led to gas leaking back through the nozzle and bubbling up into the liquid metal reservoir. Gas flow rate was therefore limited to values giving initial manifold pressures of 700 mm Hg or less. Spray distance in the chamber proved insufficient for droplet solidification, and so a tray of ethylene glycol was placed at the base of the plexiglass portion of the spray chamber to quench and collect the powder. After spraying, the ethylene glycol was poured off the powder, which was then washed, first in water and then in acetone or alcohol. After this, it was allowed to dry, examined, and sieved to obtain the size distribution.

Results

The data are presented in Table 1. In the earlier runs, manifold pressures above 760 mm Hg could not be measured, and no values are therefore given. Higher gas flow rates, although giving improved powder fineness, also led to lower metal spray rates. For the same nozzle design, lowering the gas flow to drop the initial manifold pressure from 700 mm Hg to 500 mm Hg gave a much increased metal spray rate but a correspondingly coarser powder. When powder fineness, expressed as the wt % -100 mesh, is plotted against gas flow rate, Fig. 3, and metal spray rate, Fig. 4, it becomes clear that the correlation with spray rate is much closer. It appears that the energy input into the spray process is fairly constant and is nearly independent of the volume of gas passing into the nozzle. Compare, for example, the runs numbered 10 and 13, or 9 and 17, where the spray rates and particle finenesses were very similar but the gas flow rates differed widely. A point of practical significance is that run No. 14, where the nozzle had only two gas inlet holes, gave a powder equivalent in fineness to the best produced with nozzles with 8 or 16 holes. This indicates that a quite simple nozzle is satisfactory, an important factor when designing nozzles for making powder from higher melting-point alloys.

The powder produced was spherical in shape, with very few joined spheres or large spheres with smaller 'satellite' ones attached. Sieve analyses from some of the more successful runs are given in Table 2. There is seen to be a rather wide distribution of particle sizes.

The question of the optimum nozzle diameter was also investigated. To reduce the diameter from the 4.8 mm used up to this point in the investigation would have led to unacceptably low spray rates. Consequently, a nozzle with larger diameter, 9.5 mm, was made, with 16 gas inlet holes each 2.4 mm in diameter.

Gas flow rates were high, around 100 and 80 standard litres per minute (SLM) respectively before and during the spray. Metal spray rates were also high, around 1300 g/sec. The powder produced was very coarse. It was concluded that the maximum useful nozzle diameter is significantly less than 9.5 mm, probably on the order of 6 mm.

Conclusions

The plexiglass/Wood's metal system has shown that:

a) Reasonably fine powder of good quality can be produced with this technique.

b) Nozzle diameter should not exceed 6 mm.

c) Pressure in the manifold must be adjusted, through control of the gas flow rate, to be a little over atmospheric during the spray.

d) A fairly simple nozzle design with two gas inlet holes diametrically opposed to one another should prove satisfactory.

ATOMIZATION OF CAST IRON

Apparatus and Procedure

The principles established in the model system were used in designing a unit for the vacuum-spray atomization of higher melting-point alloys. Cast iron was chosen as the alloy to be atomized. It is readily available, inexpensive, and can easily be superheated by several hundred degrees Celcius. If the cast iron was successfully atomized, it was intended to go on to more practical alloys such as steels, or Ni- and Co-base alloys. These have narrower freezing ranges and are less easily superheated, and would, therefore, be more susceptible to nozzle blockage.

A schematic drawing of the tundish and nozzle for spraying cast iron is shown in Fig. 5. The nozzle was fitted onto a steel base-plate, on which was mounted a refractory-lined tundish to hold about 6 kg of molten iron. The base-plate was designed to fit onto the top of a small pilot-plant size degassing unit. A cylindrical steel shell with a conical base, about 1.0 m in diameter by 1.5 m high, was made to fit inside the vacuum chamber to collect the powder. The nozzle itself consisted of a short piece of tubing, with gas inlet holes in its walls, and was cemented into place at the base of the tundish. The tube material had to withstand considerable thermal shock. The gas flow would keep it initially cold, despite tundish preheating, and it would then suddenly come into contact with the molten metal poured into the tundish.

The tundish was preheated with a gas torch before being installed on the vacuum chamber. Iron temperature before pouring into the tundish was 1600°C, to give some 400°C superheat.

Early Vacuum-Spray Runs

The first test was done with a nozzle tube made from 'ZRBSC-M', a refractory material developed by the Norton Company, with good resistance to thermal shock and chemical inertness in the presence of molten iron and slags. The tube was 4.0 mm in inside diameter, and had two 2.4-mm gas inlet holes. This nozzle blocked instantly, with no metal reaching the collector. To avoid blocking, a nozzle tube with greater inside diameter made of material with lower thermal conductivity was tried. It consisted of a mullite tube, 5.9-mm inside diameter, again with two 2.4-mm diameter inlet holes. Initial manifold pressure was set at 630 mm Hg, at an argon flow rate of 87 SLM. This nozzle sprayed successfully, but the product was very coarse and formed a sinter on the collector surface. After the run, the nozzle tube was found to have sheared off at the level of the inlet holes, probably from thermal shock.

Graphite, a material with greater resistance to thermal shock, was tried next. To reduce chilling in the nozzle from the steel base plate, an alumina tube was installed around the lower end of the nozzle, below the gas manifold. This was retained for all subsequent tests. An initial manifold pressure of 600 mm Hg and argon flow rate of 68 SLM were used. Nozzle diameter was 4.8 mm, with two 2.4-mm diameter gas inlet holes. During the spray, the manifold pressure rose to around 760 mm Hg and the gas flow rate remained nearly constant. The powder produced was coarse. Only the first material to reach the collector was in powder form, with the rest forming a layer of sinter. Sieve analyses were done on this first powder to form, any pieces of sinter being first removed. About 10% of the powder from this run was -100 mesh. Examination of the nozzle after spraying showed that the region below the inlets was badly eroded, the diameter being increased to about 7 mm at the exit. In a second run, a refractory wash was coated on the inside surface of the nozzle, but erosion still occurred.

To eliminate erosion while retaining good resistance to thermal shock, the original ZRBSC-M nozzle tube was bored out to 4.8-mm diameter and re-used. Initial manifold pressure was increased to 700 mm Hg. During the run, the manifold pressure rose to between 870 and 970 mm Hg, with the gas flow rate fairly steady at 57 SLM. The spray rate was much reduced from previous runs, the time to spray 4.5 kg of iron being around 25 sec. The resulting powder, however, was still quite coarse, with 16% -100 mesh.

Vacuum-Spray Runs with Silica-Glass Nozzle Tubes

The next series of trials was done with silica-glass as the nozzle tube material. Inside diameter was 5 mm, with 1-mm wall thickness. There were again two 2.4-mm diameter inlet holes. The first run, at 700 mm Hg initial manifold pressure, gave a small improvement in powder fineness, with 25% -100 mesh. A second run was done, with similar manifold pressure, to test if the greater inlet hole length and thus more directed gas jet in

the plexiglass model system was an important factor in breaking up the metal stream. A graphite ring 6.4-mm thick with two diametrically opposite 2.4-mm diameter holes was fitted over the silica nozzle tube with the holes in the ring lined up with the gas inlet holes. The wt % -100 mesh in the resulting powder was again 25, showing that the gas inlet hole length was not a critical parameter.

To increase the energy input per unit mass of sprayed metal, the nozzle diameter was reduced to 4 mm. Initial manifold pressure was 700 mm Hg. During the spray the manifold pressure rose to 920 mm Hg at a gas flow rate of 41 SLM. These conditions are similar to those for the more successful runs with the model system, see Table 1. Powder fineness was improved, with 37% -100, but this was still far inferior to that needed for a viable powdermaking process. In a further test, a 5-mm nozzle tube with four 2.4-mm diameter inlet holes was tested. The per cent -100 mesh powder rose to 48. In all these runs, the material sprayed in the later stages formed a sinter on top of the initial powder.

Figure 6 shows the bottom surface of the nozzle assembly after one of these runs. Metal had deposited around the exit, and it was postulated that this build-up could have caused interference in the spraying process to give coarsening. A further series of tests was run in which the nozzle tube extended beyond the bottom surface of the steel tundish base-plate by 13 mm. Blow-back problems with consequent nozzle blockage were increased, but the deposition of metal at the end of the nozzle tube was largely eliminated. Powder fineness did not improve.

Powder fineness for the cast iron runs as a function of metal spray rate is included in Fig. 4. It is seen that, in all cases, the cast iron gave a far coarser powder at equivalent spray rates. There are three possible explanations for this, namely:

1) There was some critical but unknown factor in the design changes made for the cast iron runs that was causing powder coarsening.

2) The cast iron droplets were in some way re-combining in the spray chamber before solidifying at the base of the collector.

3) Wood's metal is intrinsically easier to atomize than is cast iron, probably through a lower surface tension (the surface tension for Wood's metal is not known).

To elucidate these points, the refractory tundish/silica nozzle set-up was used to spray Wood's metal. With two 2.4-mm diameter gas inlet holes in a 5-mm diameter nozzle tube, the Wood's metal was sprayed at a manifold pressure of 920 mm Hg and a gas flow rate of 72 SLM. It gave a powder with 87% -100 mesh. It seems therefore that there is nothing wrong with this nozzle design, and either explanation (2) or (3) must hold. Most of the powder examined is spherical, with little agglomeration. Explanation (2) therefore seemed unlikely. This was reinforced in a further cast iron run in which the base of the collector was filled with ethylene glycol to quench any droplets that remained liquid. The powder produced was similar to that collected with the glycol. Explanation (3) would therefore seem to hold.

To examine the effects of a different material on the system, and also to see if an alloy with a shorter freezing range and less superheat could be atomized, runs with nickel were tried. With the procedures used for cast iron, the nozzle always blocked as soon as the liquid metal was poured into the tundish. To improve the preheat, the tundish was heated after being positioned on the vacuum degasser tank. The nozzle ran for a few seconds and then blocked. It was concluded that to work with nickel-based alloys or with steel, the nozzle would need a built-in heater.

The vacuum-spray trials were abandoned at this stage.

DISCUSSION AND CONCLUSIONS

The vacuum-spray procedure as developed here would work efficiently only for low-melting-point alloys that have low surface tensions. To adapt it for use with steels or superalloys, a built-in nozzle heater would be needed to prevent the nozzle blocking, which would cause a considerable complication in design. The question of how large the vacuum-spray chamber would have to be to allow solidification of the droplets has not been answered here. Those used in Wentzell's dissolved gas process were huge, but the gas jets used with the current technique should give more cooling and therefore allow the use of smaller chambers. The vacuum chamber would, however, clearly have to be larger than that of the vacuum degasser used in this work. The device would likely give better powder if blow-back into the tundish could be prevented, thus allowing a greater gas flow. At present this factor limits the gas manifold pressure to no more than around 900 mm Hg. The only way to prevent blow-back would be to apply pressure to the top surface of the reservoir. This would also allow a narrower nozzle to be used for the same metal spray rate. Again, however, the design would become more complex.

To summarize, while the technique described here could form the basis for a successful and sophisticated powder-production process, the original objective of developing a relatively simple vacuum-spray technique for the production of clean powder with low gas content could not be realized.

REFERENCES

1) Wentzell, J.M. J. Vac. Sci. Technol., 11, 169-171, 1974.

2) Olsson, R.G. and Turkdogan, E.T. J. Iron and Steel Inst., 211, 1-8, 1973.

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Table 1. Data for plexiglass/Wood's metal vacuum-spray apparatus

Run No.	Nozzle Design			Spray Conditions				Results	
	Number of Gas Inlets	Gas Inlet Hole Diam, mm	Nozzle Extension Length, mm	Manifold Pressure, mm Hg		Gas Flow, SLM*		Metal Spray Rate, g/s	% of Powder -100 mesh
				Before Spray	During Spray	Before Spray	During Spray		
1	16	1.2	7	500	>760	45	48	-	49
2	16	1.2	22	500	"	36	19	108	51
3	16	1.2	7	700	"	60	58	54	85
4	16	1.2	22	700	"	58	54	51	71
5	8	1.2	0	500	"	28	21	212	15
6	8	1.2	8	500	"	27	21	154	14
7	8	1.2	18	500	"	26	23	141	29
8	8	1.2	0	700	"	45	41	114	40
9	8	1.2	8	700	"	38	30	92	64
10	8	1.2	18	700	"	36	40	65	70
11	8	2.4	8	500	813	38	34	133	52
12	8	2.4	8	700	840	49	53	47	75
13	4	2.4	8	700	920	57	48	70	69
14	2	2.4	8	700	>920	46	38	72	76
15	1	2.4	8	700	920	21	21	162	33
16	1	3.2	8	700	894	39	35	129	41
17	1	4.8	8	700	920	62	56	86	59

*Standard litres per minute.

Table 2. Sieve analysis on Wood's metal powder made by the vacuum-spray technique

Mesh		Per cent by weight, Run No.				
		3	4	10	12	14
	+80	12.0	22.2	22.6	18.3	18.0
-80	+100	3.4	7.3	7.1	6.5	6.2
-100	+150	13.1	16.3	20.3	18.3	21.7
-150	+200	12.7	10.9	19.4	12.9	18.2
-200	+250	9.4	7.1	7.5	6.9	5.5
-200	+325	15.0	11.5	9.0	12.7	11.2
-325		34.4	24.7	14.3	24.7	19.3

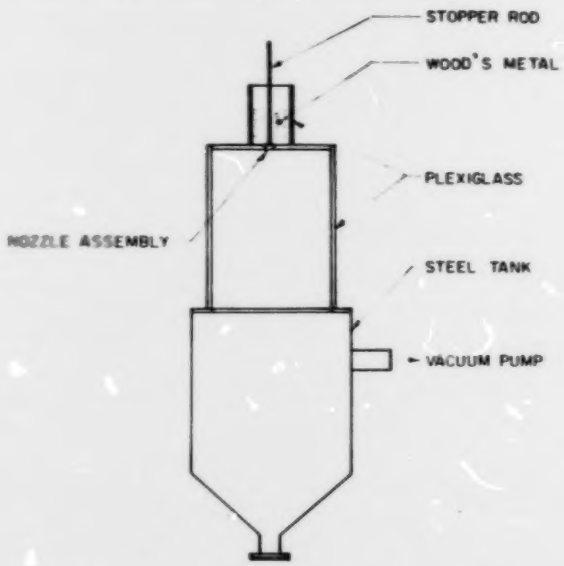

Fig. 1. The plexiglass/Wood's metal vacuum spray unit. Over-all height (neglecting the metal container), 0.92 m. Diameter of the steel tank, 0.61 m.

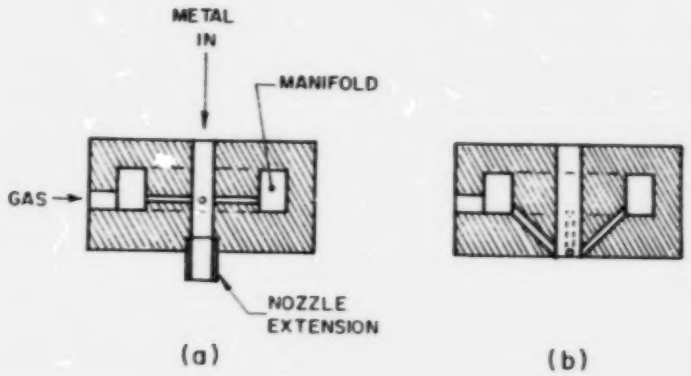

Fig. 2. Plexiglass nozzle designs. Nozzle depth 38 mm, gas manifold inside diameter 25 mm.

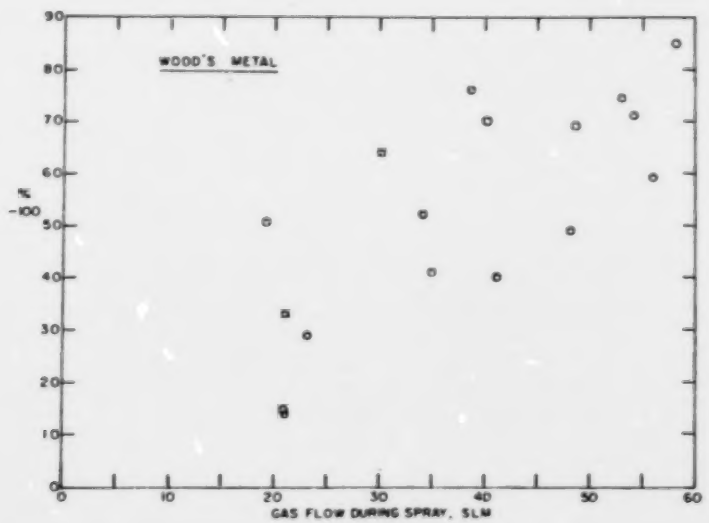

Fig. 3. Powder fineness, expressed as per cent -100 mesh, as function of argon gas flow rate (flow rate unit - standard litres per min).

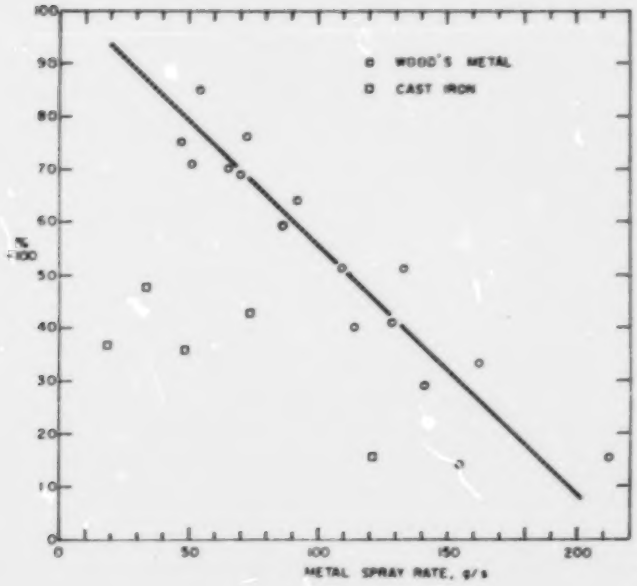

Fig. 4. Powder fineness, expressed as per cent -100 mesh, as a function of metal spray rate.

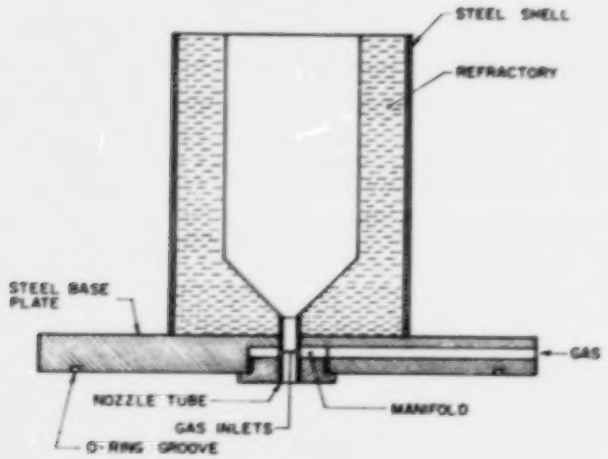

Fig. 5. Nozzle and tundish for spraying higher melting-point metals. Tundish inside dimensions, 102-mm diameter by 180 mm deep. Steel base-plate fits onto vacuum chamber of commercial vacuum degassing unit.

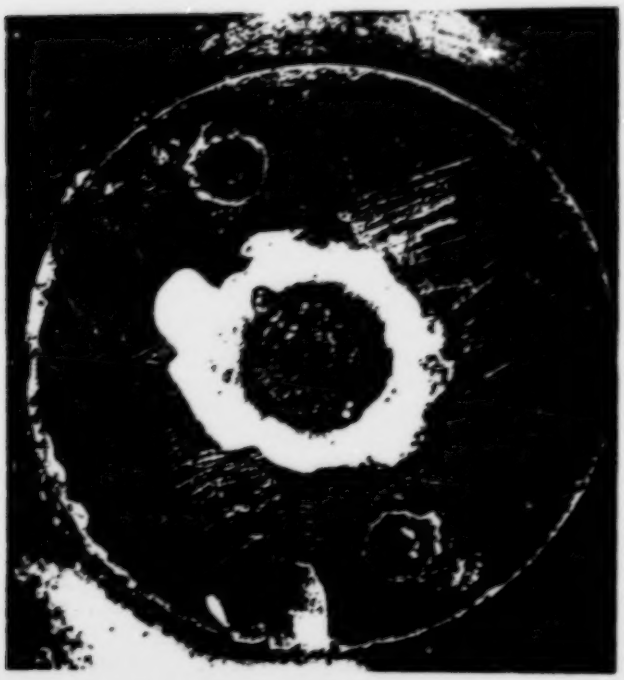

Fig. 6. Bottom view of spray nozzle assembly after cast iron spray. Silica-glass nozzle tube.